Reduction of the Superfluid Density in the Vortex-Liquid Phase of $Bi_2Sr_2CaCu_2O_y$

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In-plane complex surface impedance of a $Bi_2Sr_2CaCu_2O_y$ single crystal was measured in the mixed state at 40.8 GHz. The surface reactance, which is proportional to the real part of the effective penetration depth, increased rapidly just above the first-order vortex-lattice melting transition field and the second magnetization peak field. This increase is ascribed to the decrease in the superfluid density rather than the loss of pinning. This result indicates that the vortex melting transition changes the electronic structure as well as the vortex structure.

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The electronic structure of high-T_c superconductors (HTSC's) in the mixed state attracts much attention. In conventional superconductors (CSC's) with s-wave gap, quasiparticle (QP) states in the mixed state are localized in the vortex cores where the superconducting gap is suppressed. The QP excitation spectrum inside the vortex core consists of quantized energy levels separated by $\Delta E \sim \Delta_0/k_F\xi$, where Δ_0 is the bulk gap, k_F is the Fermi momentum and ξ is the coherence length [1]. Since ΔE in CSC's is much smaller than the scattering energies, the vortex core can be regarded as a normal metal. In HTSC's, above picture can not be applied because of the following reasons. First, the symmetry of the superconducting gap in HTSC's is not an s-wave but is most likely a d-wave . Since the amplitude of the d-wave gap is zero at the node, QP's are not localized in the vortex core but extend along the node direction [2,3]. Calculations based on the Bogoliubov-de Gennes equations suggest that there are no truly localized states in the vortex core of *pure d*-wave superconductors [4,5,6]. Secondly, even if there are localized QP states inside the vortex core owing to the mixing of the different gap symmetry [5,6,7], ΔE may exceed any other energy scales, since $k_F\xi$ of HTSC's is very small. Therefore the vortex core in HTSC's should be different from that of CSC's in either cases. Finally, in HTSC's, the greater part of the mixed state is a vortex liquid phase which is practically lost in CSC's. It may be possible that the difference in the vortex structure brings the difference in the electronic state.

Experimentally, scanning tunneling spectroscopies [8,9] and thermal conductivity measurements [10] have been used to study the QP state of HTSC's in the mixed states. High frequency surface impedance $Z_s = R_s + iX_s$ also provides useful information on the electronic state. Using high enough frequencies, one can be free from the vortex pinning and the information not only on QP's but also on superfluids can be deduced from R_s and X_s, respectively. So far, a vortex flow resistivity and a QP lifetime inside the vortex core were discussed from the R_s measurements in $YBa_2Cu_3O_y$ [11,12]. Recently, Mallozzi *et al.* measured the complex resistivity in the mixed state of $Bi_2Sr_2CaCu_2O_y$ and argued a possible d-wave effect [13]. However, measurements of Z_s near the vortex-melting transition are still lacking. In this paper, we report both R_s and X_s measurements on a $Bi_2Sr_2CaCu_2O_y$ single crystal and show that the high-frequency response can not be explained in terms of simple vortex flow. Moreover, we succeeded in measuring Z_s across the first-order vortex melting transition line and found that the reactive part, X_s, increases rapidly above the transition while there was little change in R_s. This result indicates that the superfluid density or the *amplitude* of the order parameter decreases in the vortex liquid phase.

A $Bi_2Sr_2CaCu_2O_y$ single crystal was grown by the floating zone method. The as grown crystal was annealed in air at 800 °C for 3 days and was quenched to room temperature to achieve an optimum oxygen content. A superconducting transition temperature T_c defined at zero resistivity was 91 K. Prior to the surface impedance measurements, we measured the magnetic-field dependence of the local magnetization of the same crystal using the micro Hall probe magnetometry and determined the first-order vortex melting transition field from the position of the magnetization jump. Surface impedance was measured by the cavity perturbation technique with a cylindrical Cu cavity operated at 40.8 GHz in the TE_{011} mode. The sample was located at the center of the cavity which is the anti-node of the microwave magnetic field H_{rf} being parallel to the c-axis of the sample. The dimensions of the sample were $0.5 \times 0.5 \times 0.02$ mm^3, which is appreciably larger than the normal-state skin depth (~ 1 μm at 40.8 GHz). The surface resistance R_s and the surface reactance X_s can be obtained from the changes in the quality factor of the cavity and the resonance frequency, respectively. We determined absolute values of R_s and X_s from comparison with the dc resistivity and assuming that $R_s = X_s$ (Hagen-Rubens limit) in the normal state. Using this procedure, we obtained the reasonable zero-temperature penetration depth of $\sim 2 \times 10^2$ nm. Surface impedance measurements were performed with swept temperature T under field cooled conditions to
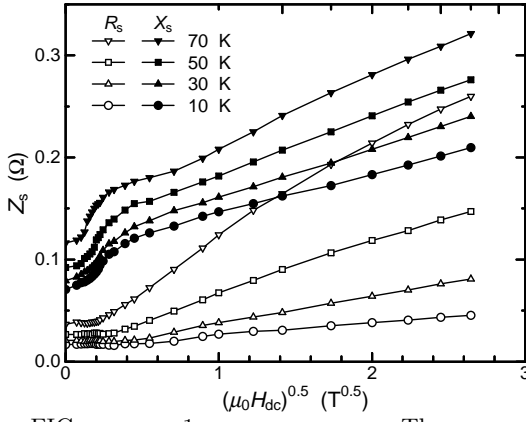
FIG. 1. The complex surface impedance in $Bi_2Sr_2CaCu_2O_y$ v.s. square-root of the applied field H_{dc}. The surface reactance X_s is larger than the surface resistance R_s, indicating that the response is reactive rather than dissipative. Note that a prominent increase in X_s at low fields.
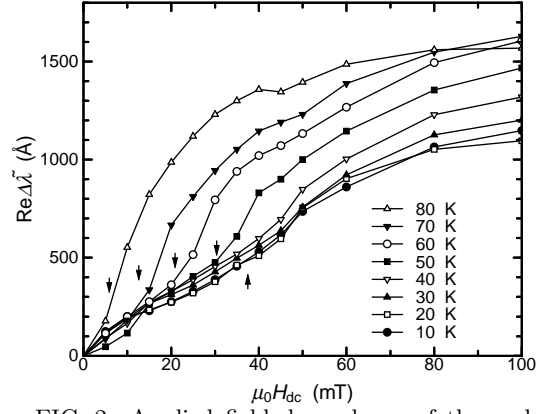

FIG. 2. Applied field dependence of the real part of the effective penetration depth. Downward arrows show the vortex melting transition field determined by the magnetization measurement. The upward arrow shows the second magnetization peak field at 30 K.

avoid any extrinsic effects associated with pinning *e.g.* giant magnetostriction [14]. In all the measurements, the static magnetic fields H_{dc} were applied along the *c*-axis.

First, we briefly introduce the general behavior of the surface impedance of type-II superconductors [15]. The surface impedance Z_s is related to the complex effective penetration depth $\tilde{\lambda}$ as $Z_s = i\mu_0\omega\tilde{\lambda}$, where μ_0 is the vacuum permeability and ω is the angular frequency. The complex resistivity $\tilde{\rho}$ is also expressed by $\tilde{\lambda}$ as $\tilde{\rho} = i\mu_0\omega\tilde{\lambda}^2$. In the Meissner state, the response is reactive and $\tilde{\lambda}$ is purely real. Therefore, $R_s \sim 0$ and $X_s = \mu_0\omega\lambda_L$ where λ_L is the London depth. In the normal state, the response is dissipative and $\tilde{\lambda}^2$ is purely imaginary. Therefore, $R_s = X_s = \mu_0\omega\delta/2$ where $\delta = (2\rho_n/\mu_0\omega)^{1/2}$ is the skin depth and ρ_n is the normal resistivity. In the mixed state, vortex dynamics affects $\tilde{\lambda}$. If the frequency is low, vortices are effectively pinned and a response is similar to that of the Meissner state except that $\tilde{\lambda}^2 \sim \lambda_L^2 + B\Phi_0/\mu_0\kappa_p$, where B is the magnetic induction, ϕ_0 is the flux quantum and κ_p is the Labusch parameter which denotes the pinning strength. On the other hand, if the frequency is high enough, the viscous loss becomes dominant and a response is similar to that of the normal state except that δ is replaced by the vortex flow skin depth $\delta_f \sim (2B\Phi_0/\mu_0\omega\eta)^{1/2}$, where η is the viscosity of the vortex motion being related to the QP excitation inside the vortex core. Within the vortex flow theory of Bardeen and Stephen [16], η is independent of B and $R_s \sim X_s \propto B^{1/2}$ at high enough fields where $\delta_f \gg \lambda_L$ and $R_s \propto B$, $X_s \sim \mu_0\omega\lambda_L$ at low fields. A crossover from the pinned regime to the dissipative regime occurs at the pinning frequency $\omega_p = \kappa_p/\eta$.

The field dependence of Z_s of the $Bi_2Sr_2CaCu_2O_y$ single crystal is shown in Fig. 1. At high fields above 0.2 T, R_s is almost proportional to $B^{1/2}$. However, X_s is larger than R_s up to the highest field we used. This result means that the contribution from the reactive parts, su-

perfluid and/or pinned vortices, can not be neglected. Therefore, the observed $B^{1/2}$ dependence of Z_s can not be attributed to the high-field region of the Bardeen-Stephen type vortex flow.

At low fields, a prominent anomaly can be seen. Above a certain field, X_s increases rapidly and saturates at a higher field while there is little change in R_s in the same field region. To examine this anomaly in more detail, we plot the field induced changes in the real part of the effective penetration depth $\mathrm{Re}\Delta\tilde{\lambda}(H_{dc}) \equiv (X_s(H_{dc}) - X_s(0))/\mu_0\omega$ in Fig. 2. At low fields and low temperatures, $\mathrm{Re}\Delta\tilde{\lambda}$ is independent of T and linear in H_{dc}. At high temperatures, $\mathrm{Re}\Delta\tilde{\lambda}$ begins to deviate from the linear H_{dc} dependence and rapidly increases above a certain field H_{kink}. In Fig. 3, H_{kink} is plotted on the magnetic phase diagram together with the vortex melting transition field H_m and the second magnetization peak field B_{sp}. Here, H_{kink} is defined at the onset of the rapid increase in $\mathrm{Re}\Delta\tilde{\lambda}(T)$ as shown in the inset of Fig. 3. The agreement between H_m and H_{kink} is excellent, indicating that the vortex melting affects $\mathrm{Re}\Delta\tilde{\lambda}$. In the present crystal, the "critical point" is located at 45 K and 33 mT. Above this field, the sharp feature in $\mathrm{Re}\Delta\tilde{\lambda}(T)$ disappears as shown in the inset of Fig. 3. However, as shown in Fig. 2, the small deviation from the linear H_{dc} dependence is still observed in $\mathrm{Re}\Delta\tilde{\lambda}(H_{dc})$ around 40 mT, somewhat higher than B_{sp}. Considering the difference between the applied field and the magnetic induction inside the sample and the different processes of the field applications in the local magnetization and the microwave measurements, it is reasonable to regard that the increase in $\mathrm{Re}\Delta\tilde{\lambda}(H_{dc})$ below 45 K and the second magnetization peak have the same origin, most likely the field induced vortex decoupling [17]. To sum up, $\mathrm{Re}\Delta\tilde{\lambda}(H_{dc})$ increases when the 3-dimensional vortex lattice model is no longer valid. Note that this anomaly only appears in the reactive response ($\mathrm{Re}\Delta\tilde{\lambda}$ or X_s) and almost no anomaly appears in the dissipative response (R_s).
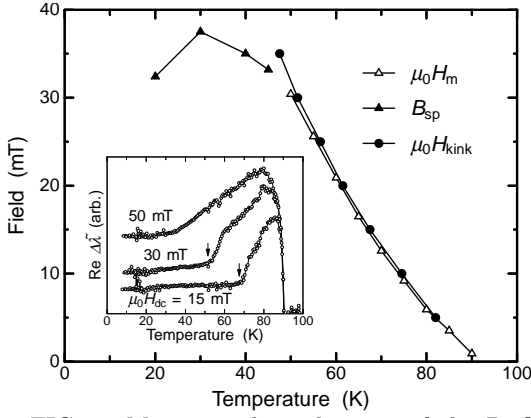
FIG. 3. Magnetic phase diagram of the $Bi_2Sr_2CaCu_2O_y$ single crystal used in this study. The kink field H_{kink} in $Re\Delta\tilde{\lambda}$, which is determined from the temperature dependence data as shown in the inset, exactly coincide with the vortex melting transition field H_m. As for the second magnetization peak field, we plot the local field B_{sp} instead of the applied field H_{sp} since the self-field becomes large at low temperatures. Above 50 K, the difference between the applied field and the local field is less than 1 mT.

Now we discuss the origin of the change in $Re\Delta\tilde{\lambda}$. Since $Bi_2Sr_2CaCu_2O_y$ does not contain magnetic ions, we can recognize that $Re\Delta\tilde{\lambda}$ consists of two components: the London penetration depth λ_L and the vortex motion. First we interpret our results in terms of the change in the vortex motion, which has been widely assumed so far [18], and will show that this scenario is inappropriate at least near the vortex melting transition of $Bi_2Sr_2CaCu_2O_y$. From the vortex-motion point of view, the increase in $Re\Delta\tilde{\lambda}$ should be attributed to the loss of pinning at the vortex melting transition. Since contributions from λ_L and the vortex motion can be separated if the response is written by the complex resistivity [15]. When the effect of vortex creep, which is important only near T_c, is neglected, the real (ρ_1) and the imaginary (ρ_2) parts of the complex resistivity are given by:

$$\frac{\rho_1}{\mu_0\omega\lambda_L^2} = \frac{s}{1+s^2} + \frac{r^2+sr}{(1+s^2)(1+r^2)}b, \qquad (1a)$$

$$\frac{\rho_2}{\mu_0\omega\lambda_L^2} = \frac{1}{1+s^2} + \frac{r-sr^2}{(1+s^2)(1+r^2)}b. \qquad (1b)$$

Here, $s = (2\lambda_L^2/\delta_{nf}^2)$, δ_{nf} is the normal-fluid skin depth, $r = \omega/\omega_p$ and $b = B\phi_0/\mu_0\omega\eta\lambda_L^2$. The parameters s and r denote the normal-fluid fraction relative to the superfluid density and the weakness of the pinning, respectively. Note that all the parameters related to the vortex motion (r and b) are included only in the second terms of the right hand sides of Eq. 1. Therefore, if only the vortex motion is considered, magnetic field dependent parts of the complex resistivity $\Delta\rho_1$, $\Delta\rho_2$ are nothing but these terms. Here, we introduce the ratio of these two terms:
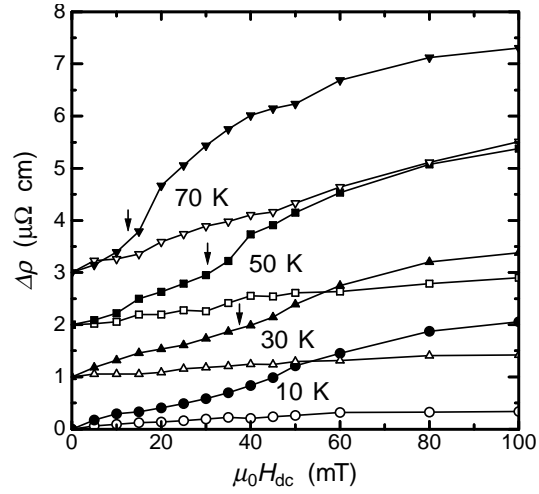

FIG. 4. Applied field dependence of the real ($\Delta\rho_1$: open symbols) and imaginary ($\Delta\rho_2$: closed symbols) parts of the field dependent part of the complex resistivity calculated from Z_s. Origins are shifted by 1 $\mu\Omega$ cm for each temperature. Arrows show the vortex melting transition fields (70 K and 50 K) and the second magnetization peak field (30 K). Note that difference between $\Delta\rho_2$ and $\Delta\rho_1$ increases above the transition.

$\Delta\rho_2/\Delta\rho_1 = (r-sr^2)/(r^2+sr)$. Let us consider the behavior of this ratio at the vortex melting transition. If the vortices melt, the effective pinning strength must be weakened. Accordingly, $r \propto 1/\kappa_p$ should increase above the transition. Since $\Delta\rho_2/\Delta\rho_1$ monotonically decreases with increasing r, $\Delta\rho_2$ should decrease relative to $\Delta\rho_1$ at the transition. This behavior is expected to be irrespective of the nature of pinning, surface or bulk, because any depinning processes cause energy losses which mainly affect ρ_1. To compare the above arguments with the experimental results, we calculated $\tilde{\rho}$ from Z_s. As shown in Fig. 4, the experimentally obtained $\Delta\rho_2$ *increases* relative to $\Delta\rho_1$ at the vortex melting transition and the second magnetization peak field. Therefore, the increase in $Re\Delta\tilde{\lambda}$ can not be attributed to the loss of pinning. Instead, it should be originated from the increase in λ_L itself. Since $1/\lambda_L^2$ is proportional to the superfluid density, this result strongly suggest that the superfluid density decreases in the vortex liquid phase of $Bi_2Sr_2CaCu_2O_y$ due to the additional pair breaking. If all the QP's are localized in the vortex core as in s-wave superconductors, the electronic state might be insensitive to the change in the vortex structure. Therefore, we speculate that the reduction of the superfluid density at the vortex melting transition is related to the d-wave superconductivity.

Next we discuss the behavior of $Re\Delta\tilde{\lambda}$ in the vortex solid phase. Since $R_s \sim 0$ in this regime, we can recognize that vortices are effectively pinned so that $r \ll 1$, and $\tilde{\lambda} \sim (\lambda_L^2 + B\Phi_0/\mu_0\kappa_p)^{1/2} \sim \lambda_L + B\Phi_0/2\lambda_L\mu_0\kappa_p$. If λ_L is independent of H_{dc}, κ_p should also be independent of H_{dc} and T, since $Re\Delta\tilde{\lambda} \propto B$ and independent of T as shown in Fig. 2. Field independent κ_p is only expected when the vortices are individually pinned by strong pin-

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ning centers. This is unreasonable because we observed the sharp first-order magnetization jump in our sample. Therefore, λ_L as well as κ_p should be H_{dc} dependent even in the vortex solid phase. Let us assume that the field dependence of λ_L is dominant, as has been argued by Mallozzi *et al.* in the vortex liquid phase [13]. The field dependent λ_L in the mixed state of d-wave superconductors is discussed in terms of the magnetic-field effect on the extended QP state by introducing the Doppler energy shift due to the circulating current around the vortices [13,19,20]. According to these theories, normal-fluid density, which is proportional to the change in λ_L, varies as $H_{dc}^{1/2}$ when $E_H > \Gamma$ and varies as linear in H_{dc} when $E_H < \Gamma$, where E_H is the averaged QP energy shift given by $E_H \sim \Delta_0 (H_{dc}/H_{c2})^{1/2}$ and Γ is the QP scattering rate by impurities or the thermal agitation. In terms of this scenario, since the observed $\text{Re}\Delta\tilde{\lambda}$ in the vortex solid phase is linear in H_{dc} and *independent* of T, $E_H < \Gamma$ and Γ should be dominated by the impurity scattering. However, the zero-field penetration depth of the same sample is found to be linear in T at low temperatures. This suggests that the dominant pair breaking mechanism should not be the impurity scattering but the thermal smearing. To resolve the discrepancy between $\text{Re}\Delta\tilde{\lambda}$ in the vortex solid phase and the zero-field λ_L, knowledge of the field dependence of κ_p and/or more detailed theoretical treatment must be needed.

Finally, we should briefly consider $\text{Re}\Delta\tilde{\lambda}$ above H_m. As shown in Fig. 2, change in $\text{Re}\Delta\tilde{\lambda}$ just above the transition is rather gradual one. Considering that the melting transition is of the first order, there should be a small discontinuity in $\text{Re}\Delta\tilde{\lambda}$ at H_m, which, we believe, is not observed because of sparse data points. The succeeding gradual change at higher fields may come from the increasing pair breaking and/or the vortex motion. If all the changes in $\text{Re}\Delta\tilde{\lambda}$ in the vortex liquid phase arise from the pair breaking [13], the quantity $\lambda_L(T=0)^2/(\text{Re}\tilde{\lambda})^2$ at low temperatures represents the superfluid fraction f_s. We calculated this quantity at 10 K and found that $\lambda_L(T=0)^2/(\text{Re}\tilde{\lambda})^2 \sim 0.3$ at $H_{dc}=1$ T. Even if d-wave effect is taken into account, this value is too small since f_s is given by $f_s(H_{dc}) \sim 1 - (H_{dc}/H_{c2})^{1/2}$ [20] and H_{c2} is considered to be around 100 T. Therefore, both the increase in λ_L and the contribution from the vortex motion should be considered in the vortex liquid phase. We stress here again that the behavior of $\Delta\rho_2/\Delta\rho_1$ at the transition can not be explained in terms of the vortex motion alone, even though the two effects contribute to $\text{Re}\Delta\tilde{\lambda}$. To separate both contributions, detailed frequency dependence measurements are indispensable. This experiment is now underway.

In conclusion, we have measured the complex surface impedance $Z_s = R_s + iX_s$ of a $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_y$ single crystal in the mixed state. We succeeded in detecting the change in Z_s at the first-order vortex melting transition and the second magnetization peak field for the first time. Above the transition, X_s which is proportional to the real part of the effective penetration depth increases while there was little change in R_s. From the analysis of the complex resistivity deduced from Z_s, we showed that the increase in X_s can not be ascribed to the loss of pinning but arises from the reduction of the superfluid density. Namely, the additional pair breaking mechanism may exist in the vortex liquid phase. Our results indicate that not only the phase but also the amplitude of the order parameter take different values in different vortex phases. We speculate that this effect is related to the d-wave superconductivity.

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